

LITHIUM SYNTHETIC REAGENTS: DIMERIZATION AND INTRAMOLECULAR ASSOCIATION.
DOUBLE BRIDGING IN "DIANIONS"

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Abstract - The dimerization energies of lithium compounds, $2\text{LiX} \rightarrow (\text{LiX})_2$, are quite large and depend principally on the electronegativity of X. Intramolecular association, the equivalent of dimerization, is comparably favorable energetically. Thus, many polyolithium compounds, including many synthetically useful "dianions", are found by molecular orbital calculations to prefer structures in which the lithium atoms bridge symmetrically. An example is *o,o'*-dilithiobiphenyl, whose predicted structure has now been confirmed by X-ray spectroscopy. Dilithiated species based on propene ($\text{CH}_2\text{CHCHLi}_2$), propane ($\text{LiCH}_2\text{CH}_2\text{CH}_2\text{Li}$), acetaldehyde (Li_2CHCHO), acetone ($\text{LiCH}_2\text{COCH}_2\text{Li}$ and $\text{Li}_2\text{CHCOCH}_2$), and amides (e.g., HCOLi_2) are among 1,3-double bridging examples. Besides *o,o'*-dilithiobiphenyl, 1,4-double lithium bridging is exhibited by 1,4-dilithiobutane, 1,4-dilithio-2-cis-butene, and 1,4-dilithio-1,3-cis,cis-butadiene. While electrostatic interactions provide the simplest explanation for these structures, multi center covalent bonding also is important in determining geometrical details and energetic differences.

INTRODUCTION

Despite their great synthetic utility, carbanions are the least well understood class of organic reactive intermediates (1). Thus, while the isopropyl cation and radical can be observed directly in condensed phases and their energies are established from gas phase measurements, the isopropyl anion is unknown and appears to be incapable of existence as an isolated entity. The methyl radical has an electron affinity of only 1.84 kcal/mol; methyl groups in the higher alkyl anions are destabilizing (2). Hence, the isopropyl anion is not expected to be bound and would eject an electron spontaneously in the gas phase.

Most synthetic intermediates referred to colloquially as "anions" because of their chemical behavior, are in reality organometallic compounds with polar character (1). Lithium compounds, with which this presentation will deal, are generally associated not only in the solid, but also in the gas phase and even in solution in coordinating solvents. The tetramerization energy of methyllithium is about 125 kcal/mol (3); even the highly coordinating ligand tetramethylethylenediamine (TMEDA) is incapable of dissociating such a tightly bound cluster (4). There is no justification in writing " CH_3Li " as a monomer; such oversimplified representations prejudice our thinking. While available X-ray structures reveal much about the complexity of "anionic" species, (5) a systematic investigation of the structures and energies of lithium compounds is only possible at present through theoretical calculations (6).

A remarkable number of organic species, loosely called "dianions" or "polyanions", also are useful synthetic intermediates (1). With the exception of a few X-ray structures on dilithium compounds (5), even less little detailed experimental information is available than for the monoanions. Energies are unknown; neither solution nor gas phase data is available. As with the isopropyl anion, most of the smaller carbodians are incapable of existence as isolated entities. Because of electrostatic repulsion, the electrons in outer orbitals will not be bound.

The metallic counterions are intimately involved in stabilizing "polyanionic" systems. The nature of these organometallic species is the main subject of this report.

Our examination takes advantage of the increasingly efficient, sophisticated, easy to use, and readily available calculational methods based on molecular orbital theory (6). Systematic studies of many smaller species at an adequate theoretical level have been carried out and permit energetic and structural comparisons to be made. Such calculations on isolated organometallic monomers are not as unrealistic as might be imagined; the effects of association and solvation often tend to be comparable for similar species. While we are evaluating association and solvation effects computationally, these results will not be

presented here. However, some useful background data may be helpful: the strength of lithium bonds and energies of association.

THE ENERGIES OF LiX BONDS

The homolytic dissociation energies of lithium bonds in monomeric LiH (56.0), Li₂ (24.1), LiOH (105.4-4), and LiF (136.3 kcal/mol) are known experimentally (7). By means of theoretical evaluations, we have estimated the energies of the lithium bonds in LiNH₂ (65-4) and LiCH₃ (45-2 kcal/mol) (8). The bonds to the more electronegative atoms or groups are stronger due to increasing ionic character. The energy required for ionic dissociation, e.g., D(CH₃-Li⁺) = 167 kcal/mol, much higher than for homolytic dissociation, is too great to be overcome by solvation in the typical n-donor solvents used experimentally. We calculate the total solvation energy for diethylether-CH₃Li monomer to be on the order of 20 kcal/mol and about 70 kcal/mol for diethylether-Li⁺. It is thus unrealistic energetically to think of methyllithium giving a methyl anion which then reacts, e.g., by adding to a carbonyl group. Instead, organometallic aggregates (e.g., (CH₃Li)₄) are the reactive species. Since the association energies are large (about 125 kcal/mol for the tetramerization of methyllithium), the average bond dissociation energies in the aggregates are much higher (e.g., about 76 kcal/mol for C-Li in the (CH₃Li)₄ tetramer).

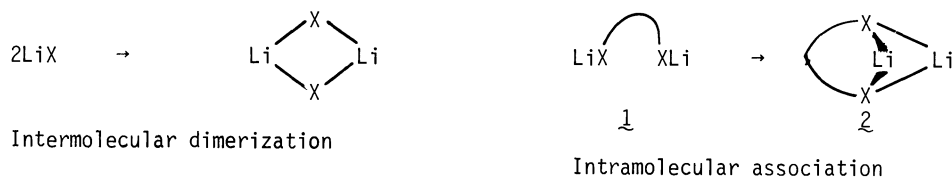
LiX DIMERIZATION ENERGIES

Only the dimerization energies of LiH, LiOH, and LiF are known experimentally. To complete the first-row (LiX) set, we have calculated all the possibilities. The results, which include some studies of conformational effects, are plotted in Figure 1 vs the electronegativities of X. A remarkable straight-line relationship is seen with only (LiOH)₂ and (LiNH₂)₂ deviating. The perpendicular conformation of the latter allows the nitrogen lone pairs to enhance stability via multicenter bonding. In the planar (LiNH₂)₂ conformation, the nitrogen lone pairs participate in a destabilizing 4 π electron interaction. Steric effects are not responsible for the downward deviation of the planar form; (LiBH₂)₂, which prefers a planar conformation (the diborane-like structure is less stable), correlates well. The oxygen lone pairs contribute to the smaller deviation of (LiOH)₂ from the correlation line. Fluorine is a much weaker π donor and (LiF)₂ is ionic (9).

Rather remarkably, a simple electrostatic calculation reproduces the correlation line shown in Figure 1 quantitatively. Such calculations are based on the ab initio optimized geometries and the assumption that positive point charges are located at the lithium positions and negative point charges at the locations of the heavy atoms, X, in each group. While the agreement of the crude electrostatic model is to some extent fortuitous, the predominant coulombic nature of the interactions in these dimers is apparent.

INTRAMOLECULAR ASSOCIATION

If lithium compounds can dimerize with the release of large amounts of energy, the intramolecular equivalent is expected. This intramolecular "dimerization" or association can be illustrated schematically by the "cyclisation" of a dilithium species, 1 → 2. In 2, lithiums form a symmetrical double bridge akin to the four-membered ring LiX dimers. Streitwieser has emphasized the electrostatic advantages in 2 of such "ion triplet" arrangements (9a). We have discovered a large number of such symmetrically doubly bridged polyolithium compounds calculationaly (10). While ionic bonding is of indisputable importance, we believe that multicenter covalent interactions may be decisive in determining many structural details. Several examples will illustrate the principals involved.



How do we carry out such computational investigations? Lithium compounds do not obey classical structural rules. Thus, potential energy surfaces must be searched widely in order to ensure that the most stable geometry (the global minimum) has been found. This cannot be done rigorously, but only by trial and error. Many structural candidates must be examined, and a thorough search is very costly in computer time. Except for very small systems, we begin with MNDO calculations (11). This semiempirical method, which has been parameterized for lithium by Thiel and Clark (12), provides a cost-efficient "first look". Unfortunately, the parameterization is compromise; while Li-O and Li-N interactions are rather accurately reproduced, Li-C bond energies are overestimated by ~ 30 kcal/mol ($\Delta H_f^\circ(\text{CH}_2\text{Li}) = -1.4$ kcal/mol (MNDO) vs the best estimate, +28 kcal/mol). Especially when different kinds of atoms are involved in a lithium molecule, relative MNDO energies of different isomers may be

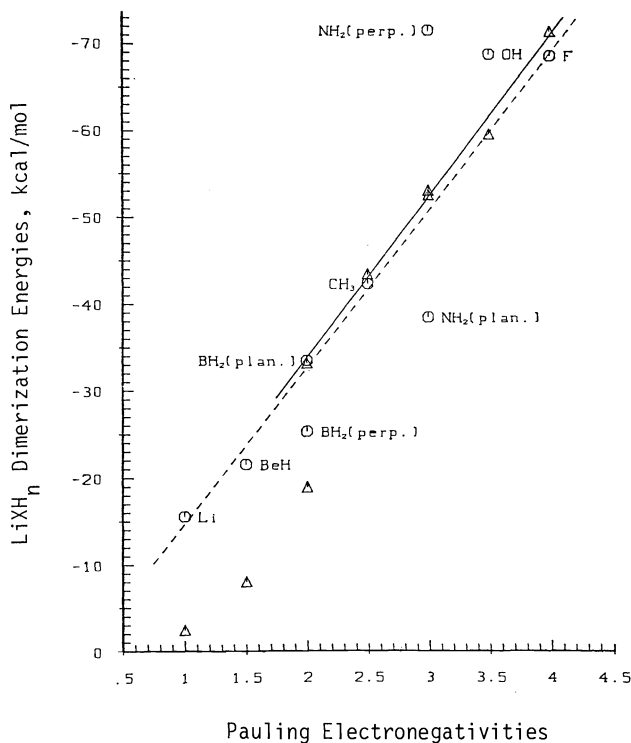


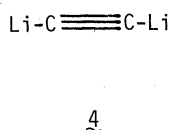
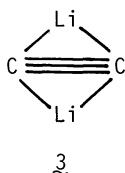
Figure 1. A plot of dimerization energies, $2\text{LiXH}_n \rightarrow (\text{LiXH}_n)_2$ (kcal/mol), vs Pauling electronegativities of the first-row elements, X. The points, \circ , from 3-21G//3-21G calculations are correlated by a dashed line (-----). For both BH_2 and NH_2 , two values, corresponding to planar and perpendicular C_{2v} arrangements are shown. The points, Δ , corresponding to data from electrostatic calculations, are plotted with a solid line (———). These calculations assume 3-21G distances between Li and X, and full positive (Li^+) and negative (X^-) charges.

incorrect. On the other hand, the automatic geometry optimization routines built into the MNDO program often reveal minimum energy structures which would tax the imagination of a conventionally-trained chemist. Used with understanding, MNDO is a powerful preliminary method.

We next select the most favorable structures for ab initio investigation. The minimal basis set, ST0-3G, has now been abandoned for this purpose; the efficient small split valence basis, 3-21G, gives more accurate geometries and energies when compared with higher level results. We next carry out the highest level calculations which are presently feasible. Fortunately, even relatively low ab initio levels tend to describe lithium compounds well because of the large number of extra basis functions present. In addition, electron correlation effects usually are not very important in ionic or highly polar molecules. Higher levels of theory are necessary for refined values, but often do not change the qualitative conclusions or the ordering of isomers. We have published several examples, $\text{C}_3\text{H}_5\text{Li}$ (10a), $\text{C}_2\text{H}_2\text{Li}_2$ (10b), and 1,2-dilithioethane (10c) isomers.

DILITHIOACETYLENE

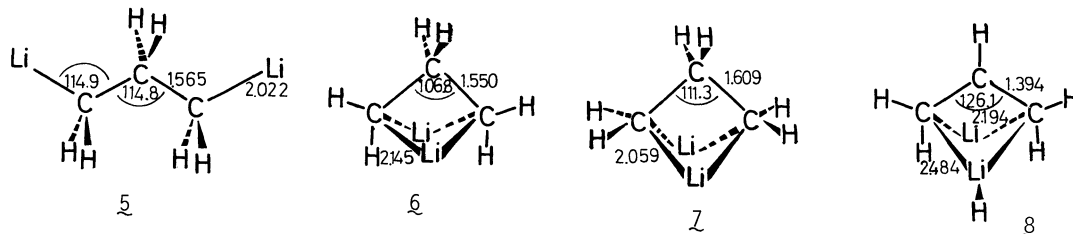
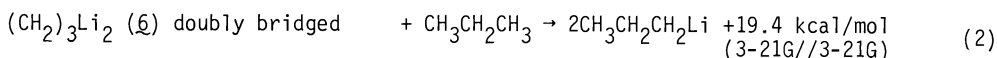
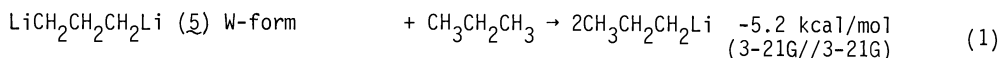
Dilithioacetylene, C_2Li_2 , which is particularly sensitive to the theoretical level employed, is an exception. ST0-3G favors the doubly bridged form, **3**, by 20.5 kcal/mol over the linear geometry, **4**. (10d) Split valence basis sets favor the linear form; the preference increases with the size of the basis set: -1.4 (3-21G//3-21G), -4.7 (4-31G//4-31G), and -5.8 kcal/mol (6-31G//6-31G). When d type polarization functions are added, the bridged form becomes more stable once again, by 7.0 kcal/mol at 6-31G**//6-31G*. Correlation increases this trend modestly, and the difference is 9.5 at MP2/6-31G* and 9.8 kcal/mol at MP3/6-31G* (10d).



1,3-DILITHIOPROPANE

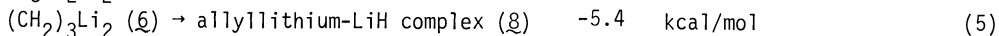
By indirect metallation procedures, 1,3-dilithiopropene has been prepared for the first time by Bickelhaupt's group (13). LiH elimination occurs readily to give allyllithium. However, 2,2-methyl-1,3-dilithiopropene, where elimination is precluded, is much more stable than ordinary primary alkylolithiums in ether solution.

Calculations indicate symmetrical double bridging to be responsible for this extra stability (14). Using the parent compound as a computational model, the extended conformation, 5, is found to be thermodynamically unfavorable relative to disproportionation (eq. 1). In contrast, the doubly bridged form is 24.6 kcal/mol (3-21G//3-21G) more stable than 5 and 6's disproportionation (eq. 2) is strongly endothermic.



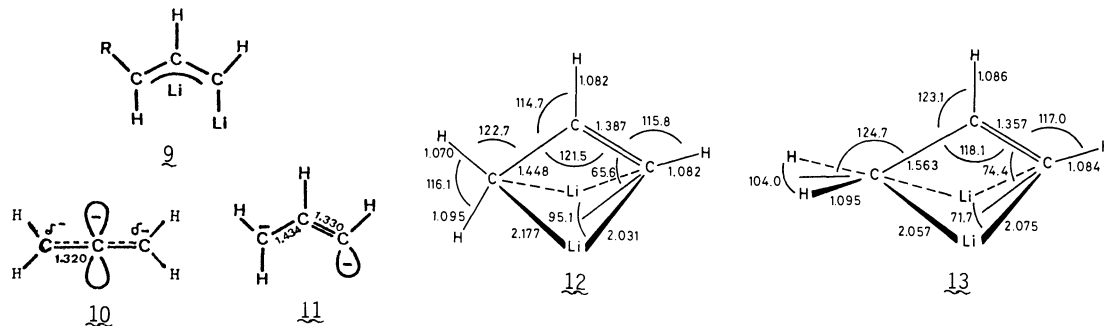
Because of the eclipsed methylene groups in 6, the structure appears somewhat unfavorable. The staggered alternative, 7, also offers a 14.7 kcal/mol electrostatic advantage over 6, based on point charge calculations. Nevertheless, 6 is 16.7 kcal/mol more stable than 7 at the 3-21G level, due to improved multicenter covalent bonding.

Why does 1,3-dilithiopropene eliminate LiH so readily? As indicated by eq. 3, complete elimination is endothermic as is the loss of LiH from n-propyllithium (eq. 4). However, if these elimination reactions proceed via lithium hydride complexes the thermodynamic driving force would be increased. The LiH-ethylene complexation energy is 12.9 kcal/mol, but the association energy of LiH with allyllithium is much larger, and the conversion of 6 to the allyllithium-LiH complex, 8, is exothermic (eq. 5).



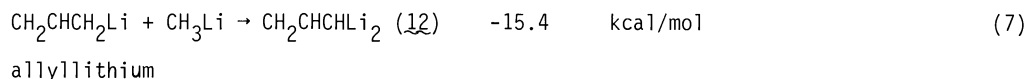
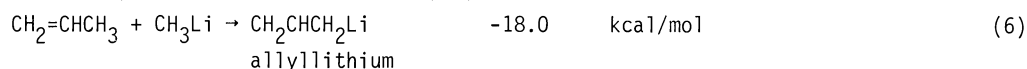
1,3-DILITHIOPROPENE

Klein and Medlik-Balan discovered that propene could be dilithiated (15). The first step, to give allyllithium, was followed by a slower dimetallation to give $\text{CH}_2\text{CHCHLi}_2$, a species originally formulated as 9. The alternative 1,2-dimetallation to give $\text{CH}_2\text{C}(\text{Li})\text{CH}_2\text{Li}$ was not observed. This is surprising because the dianion corresponding to the latter² (10) is more stable than the dianion, 11, corresponding to the observed product.



Symmetrical double lithium bridging again provides a satisfying explanation (16). Two structures, 12 and 13, are quite close in energy. The 0.7 kcal/mol less stable form, 13, has a 90° rotated CH_2 group which "shuts off" allylic resonance. However, the geometries and energies of these systems indicate both to be largely π -localized with quite unequal C-C bond lengths. In 12, electrostatic repulsion is responsible for the π localization.

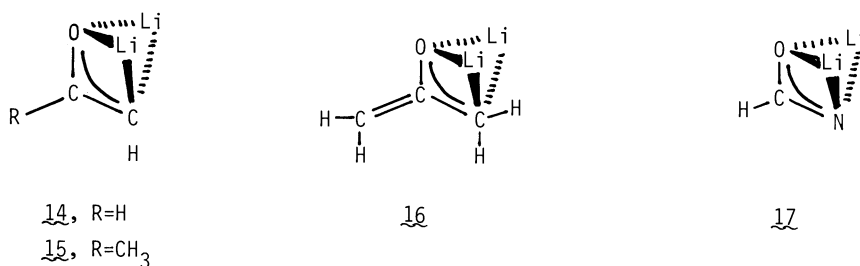
Eq. 7 and 8 model the dimetallation energetics. Consistent with the experimental observations, the first substitution to give allyllithium (eq. 6) is somewhat more exothermic than the second step (eq. 7) to give dilithiopropene.



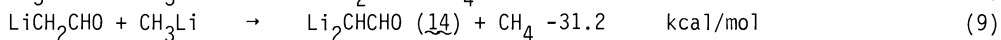
Klein, Kost, Shriver, and Streitwieser have also studied this system and come to similar conclusions (17). 1,3-Double lithium bridging is also indicated by our calculations on polyolithiated propynes and allenes, e.g., LiCHCHCHLi, C₃HLi₃, and C₃Li₄ (10a).

1,3-SYMMETRICAL BRIDGING IN HETEROATOM SYSTEMS

A remarkable number of dilithiated heteroatom species are useful synthetic reagents. Thus, ketones can be dimetallated: "1,3-dianions" have been known for some time (1a,18), and "1,1-dianions" prepared more recently (19). Carboxamides, RCONH₂, can also be metallated twice (1a). We have modeled these systems with the simplest possibilities, and have examined potential energy surfaces of the following dilithiated molecules: acetaldehyde, HCOCHLi₂; acetone, 1,3-LiCH₂COCH₂Li (18) as well as 1,1-CH₂COCHLi₂, and formamide, HCONLi₂. In all these cases, symmetrically doubly bridged lithium forms, 14-17, are found to be very stable (20).



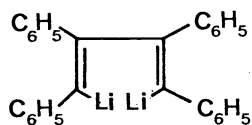
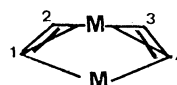
Energetic relationships involving these species are summarized in eqs. 8-12 using 3-21G data. As expected, the first lithiation (eqs. 8 and 10) is more exothermic than the second (eqs. 9 and 11). However, the second metallation steps also are very favorable thermodynamically. This would not be the case if gegenion stabilisation were not present. In other words, the corresponding free dianions would not enjoy favorable thermodynamics. Also as expected from experiment (18), 1,3-dilithiation of acetone is more favorable than 1,1-dilithiation. Alternative forms of 1,3-dilithioacetone in which each lithium bridges a different carbon and oxygen are found to be less stable than 16.



Many more known systems accord the possibility of 1,3-, 1,4-, and higher symmetrical double lithium bridging. In the literature, structural representations of lithium compounds are employed which reflect the expectations of classical bonding. However, lithium almost never favors the same location in a molecule as does hydrogen, and one can almost be certain that classical representations of the structures of lithium compounds are incorrect. Instead, the possibility that polymetallated compounds involve symmetrical lithium bridging should be considered. Such formulation may help to explain the ready formation and the behavior of such systems.

1,4-CIS,CIS-DILITHIOBUTADIENE

When diphenylacetylene is reacted with lithium metal, dimerization occurs to give the dilithiated tetraphenylbutadiene traditionally depicted by 18. Even though the lithiums would appear to be sterically and electrostatically uncomfortable in 18, this representation is employed because ready ring closure reactions to give five membered rings can be realized by a variety of reagents. Besides 18, other 1,4-dilithiobutadiene systems are known; the lithiums invariably are oriented cis,cis (21).

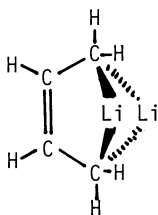
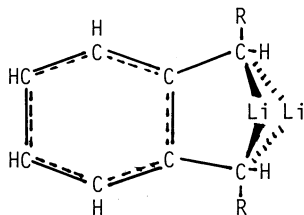
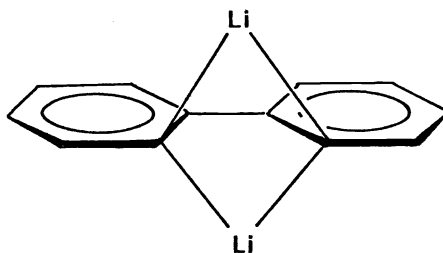
18192021, M=Li

By modelling these systems computationally with various 1,4-dilithiobutadiene geometries, e.g., 19-21, we have shown the symmetrically doubly bridged structure, 21, to be by far the best (21). While favorable electrostatic interactions in 21 undoubtedly contribute greatly to stabilization, 4 π electron aromaticity, Möbius-Hückel in character, also is significant. Because of the bridging lithium atoms, the two occupied π orbitals are different in character. One, with no phase inversions, is of Hückel type; the other has one phase inversion not passing through nuclei and is Möbius in character.

1,4-DILITHIO-CIS-BUTENE

It is appropriate to end this presentation with two examples for which recent X-ray structural evidence is available. Some years ago, Bates' group in Arizona found that 1-butene as well as the 2-cis and 2-trans-butene could be dimetallated to give the "butadiene dianion" (1c). Computational studies, both at MNDO and 3-21G, indicate the symmetrically doubly lithium bridged cis-isomer, 22, to be more stable than all other geometries, trans and cis, we have considered (20). Rotation by 90° of one or both CH₂ groups in 22 is unfavorable energetically. This indicates the importance of cyclic 6 π electron delocalization, the ring being completed by the bridging Li's. The 3 π MO's have Hückel, Möbius, and Hückel character.

While the X-ray structure of 22 has not been determined, that of a not too distant relative has recently been reported by Lappert, Raston, Skelton, and White (22), a α, α' -dilithio-oxylylene derivative, 23, additionally stabilized by α -Si(CH₃)₃ groups. The crystal structure shows symmetrical 1,4-dilithium bridging clearly. Each lithium atom additionally is solvated by a tetramethylethylenediamine (TMEDA) ligand.

2223, R = Si(CH₃)₃24

o,o'-DILITHIOBIPHENYL

Many neighbouring groups of diverse character "activate" certain C-H bonds towards metallation (1). A lithium substituent, already present in a molecule, often does this particularly well, and directs dilithiation to a particular position. W. Neugebauer has shown recently that biphenyl can be dilithiated directly in a synthetically useful procedure to give o,o'-dilithiobiphenyl, 24 (23). That this reaction occurs in a stepwise manner, was shown by the preparation of o-lithiobiphenyl separately, and demonstrating that further metallation is directed to the opposite ring. Noting the structural similarity between 1,4-cis,cis-dilithiobutadiene (18) and o,o'-dilithiobiphenyl, we postulated a doubly bridged structure 24 for the latter compound (21,23). MNDO calculations supported this suggestion (23), but the molecule is too large for an ab initio examination.

We are now pleased to report an experimental verification of our prediction of symmetrical 1,4-lithium double bridging. This is shown in the X-ray structure determined by U. Schubert, München, recently (24). TMEDA solvation again is present.

CONCLUSIONS

Symmetrical double bridging by metals is a ubiquitous structural feature of "polyanion" systems. Alkali and other metals, besides lithium, are likely to behave similarly. Such double bridging is favored electrostatically, and may also benefit from multicenter covalent bonding. Synthetic chemists are well advised to consider such structural possibilities for their reactive intermediates and interpretations of chemical behavior.

ACKNOWLEDGMENTS

Many coworkers, cited in the references, carried out the work discussed here. The principal contributors were Dr. A.J. Kos, J. Andrade, E. Kaufmann, and W. Neugebauer. Much of our program on the nature of lithium compounds is a collaborative effort with John A. Pople, Carnegie-Mellon University. We are also grateful to Professor Pople and his group for the development of the GAUSSIAN series of programs. We thank W. Thiel and T. Clark for the lithium MNDO parameterization and the Deutsche Forschungsgemeinschaft and the Fonds der Chemischen Industrie for financial support.

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